

Superconductivity in CuCl/Si: possible excitonic pairing?

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The search for superconductivity with higher transition temperature (T_C) has long been a challenge in research efforts ever since its first discovery in 1911. The effort has led to the discovery of various kinds of superconductors and progress in the understanding of this intriguing phenomenon. The increase of T_C has also evolved; however, the dream of realizing room-temperature superconductivity is far from reality. For superconductivity to emerge, the effective quasiparticle interaction should overcome the repulsive Coulomb interaction. This can be realized via lattice or spin degrees of freedom. An alternative pairing mechanism, the excitonic mechanism, was proposed 50 years ago, hoping to achieve higher T_C than by phonon mediation. As none of physics principles has ever prevented excitonic pairing, the excitonic pairing mechanism is revisited here and we show that the effective quasiparticle interaction without lattice and spin can be attractive solely electronically.

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For superconductivity to emerge, the formation of Cooper pairs is necessary, which requires an attractive quasiparticle interaction. The Cooper pair is a bound state of two electrons. Two electrons on the Fermi surface with momenta \mathbf{k} and $-\mathbf{k}$ scatter to other states with momenta \mathbf{k}' and $-\mathbf{k}'$ via an attractive interaction, which is commonly called a binding glue. The thermodynamic properties of a superconductor greatly depend on the kind of the glue, for example phonons or spin-fluctuations.

According to BCS theory[1], the superconducting transition temperature (T_C) is expressed as

$$T_C \approx \Theta \cdot \exp(-1/\lambda), \quad (1)$$

where λ is the effective pairing strength, and the prefactor Θ is the characteristic energy scale of mediating bosons. In phonon-mediated superconductors, Θ is the Debye temperature (Θ_D), and λ arises from the electron-phonon coupling constant; many attempts to raise λ to obtain a higher T_C often resulted in a lattice instability (as in A15 compounds[2]). Materials with high Θ_D usually have low λ . At a time it was believed that $T_C \lesssim 30\text{K}$ by phonon mediation[3]. Pairing mechanisms other than phonon with larger Θ has been pursued in this context.

Pairing by the spin-fluctuation mechanism relies on spin-dependent interactions, whose details are very sensitive to the crystal structure and to the magnetic properties[4, 5]. Despite the progress in several decades, and hope that a spin-fluctuation mediation might offer a better mean than phonons for achieving higher T_C , an exact formulation of the spin-fluctuation has not been completed at the level of BCS or Eliashberg theory[6]. Either phonon- or spin-fluctuation mediated, superconductivity requires an attractive effective interaction, which, in principle, yields a gap equation that can be

solved for T_C . This, however, is a rather formidable task. Instead, in the framework of the BCS approximation, the quasiparticle interaction is treated as an attractive potential well for frequencies less than Θ_D ($\Theta_D \ll \omega_F$), while the Coulomb repulsion prevails over the entire frequency range, as illustrated in Fig. 1(a). In the excitonic pairing, on the other hand, another attractive potential is added for frequency less than Θ_E ($\Theta_D \ll \Theta_E \ll E_F$), as shown in Fig. 1(b)[7]. Before we proceed, a brief history of the excitonic mechanism is first given.

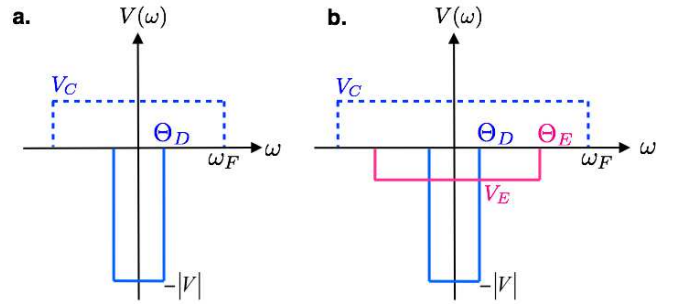


FIG. 1. (Color online) Schematic illustration of the effective potential in (a) BCS and Eliashberg theory, and (b) excitonic pairing (or ABB model). Θ_D and ω_F are the Debye frequency and the Fermi energy, respectively, $V_C = 0$ in BCS theory. Θ_E is the characteristic energy scale of excitons.

History of excitonic mechanism. In the early 1960's, Little[8] and Ginzburg[9] proposed an alternative pairing mechanism, an excitonic mechanism, where the mediating bosons were excitons or longitudinal plasmons, which have a much higher characteristic energy scale than phonons, so that T_C can be greatly enhanced due to the larger prefactor

in Eq. (1). Specific geometries were proposed: (i) a linear chain with a polarizer alongside [Little], (ii) a layered system with an insulator-metal-insulator *sandwich* structure [Ginzburg]. Later, Allender, Bray, and Bardeen (ABB) studied a metal-semiconductor interface more in detail[7]. ABB showed that excitonic mediation can enhance T_C without excluding the phonon mediation. In ABB theory, Cooper pairs in the metallic region penetrate into the semiconducting gap region and scatter back to the metallic region, during which virtual excitons are created and annihilated. The potential well in ABB theory has an additional attractive part, $V_e < 0$ with cutoff frequency Θ_E much larger than Θ_D , as is schematically illustrated in Fig. 1(b). Inkson and Anderson (IA) objected to the ABB theory, arguing that the effective interaction is of no help in pairing[10]. However, in reply to IA, ABB questioned the validity of the dielectric function that IA used[11]. Subsequently, Cohen and Louie (CL) considered the ABB theory from IA's perspective[12]. CL also claimed that IA's dielectric function is too approximate and argued that an attractive interaction could exist. Using a rather simplified geometry, however, these authors could not conclude that there is pairing. Both ABB and CL insisted that the inclusion of local-field effects would clarify the debate between ABB and IA. Zakharov *et al.* extended CL's work to a Si-jellium-Si sandwich structure with rigorous *ab initio* calculations of the dielectric function and showed that an attractive interaction could exist for certain frequencies and wave vectors but there is no overall pairing[13]. However, the introduction of jellium is rather artificial, which can be different from the metallic state arising at the interface.

On the experimental side, following ABB, several efforts were made, especially using the interface of Pb-based narrow gap semiconductors[14, 15]. This turned out to be fruitless despite some indications of superconductivity. In the late 1970's, possible superconductivity in CuCl under hydrostatic pressure had attracted the physics community, which exhibited diamagnetism over 90 K[16]. To explain possible superconductivity in CuCl, Abrikosov proposed "metallic excitonium", an analogue to the electron-phonon mediation for $m_h/m_e \gg 1$: The hole replaces the role of the nucleus, and Cooper pairs are mediated via the exchange of the metallic excitonium giving a higher T_C than with phonon mediation. However, Abrikosov's idea was based on the incorrect belief at that time that CuCl had an indirect band gap. Despite numerous subsequent works, superconductivity in CuCl remains speculative[17, 18]. Just before the discovery of superconductivity in cuprates[19], another possible superconductivity was reported in the CuCl/Si [111] interface with diamagnetism in the range of 60–150 K, an anisotropic magnetic response, and a resistivity drop by five orders of magnitude at 77 K[20, 21]. However, the works on CuCl/Si was not followed up for long time and the superconductivity of CuCl/Si still remains open[22].

Rather recently, superconductivity at an interface has been reported in chalcogenides[23, 24] and oxides[25], although the pairing mechanism is not clear yet. On the theoretical

side, the possibility of the excitonic pairing has been raised in metal-halide interfaces[26] and microcavities[27]. With advances in fabrication techniques, and increasing interest in interface phenomena, revisiting superconductivity with excitonic pairing in CuCl/Si would be of great interest.

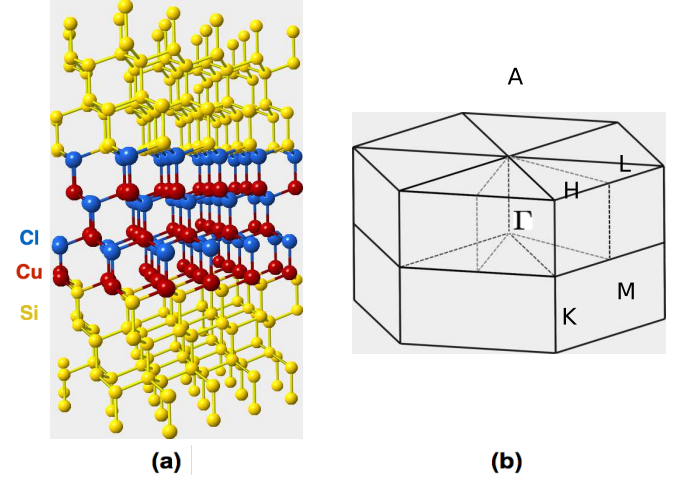


FIG. 2. (Color online) (a) Structure of the CuCl/Si [111] superlattice for $n = 3$, where Cu, Cl, and Si atoms are denoted by red, blue, and yellow spheres, respectively. (b) Brillouin zone at high symmetry point directions.

Electronic Structure. Density functional calculations are performed using the highly precise full-potential linearized augmented plane wave (FLAPW) method[28] with the local density approximation (LDA) for the exchange-correlation potential by Hedin and Lundqvist parametrization[29]. Muffin-tin (MT) radii of 2.15 a.u. (Cu) and 2.00 a.u. (Si and Cl) are used, where the angular momentum expansion $\ell \leq 8$ is used inside the MT spheres. Cutoffs for basis functions and potential representation are 7.22 htr and 103.68 htr, respectively. Structural optimization is done with a force criteria of 0.05 eV/Å, where the *in-plane* lattice constants are taken from experiments. For summations in the Brillouin zone, a $12 \times 12 \times 4$ mesh is used for self-consistent calculations, and $48 \times 48 \times 24$ mesh for Fermi surface plots. Results do not change when a denser k points mesh is used.

CuCl and Si crystallize in the zincblende and the diamond structure with lattice constants, 5.41 Å and 5.43 Å, respectively, giving a very small lattice mismatch in the CuCl/Si superlattice. The [111] direction of both the zincblende and the diamond structure are hexagonal with an *ABC* stacking sequence. To ensure a periodicity along the *c* direction, $(\text{CuCl})_n/\text{Si}_{4n}$ is taken into account, where n denotes the number of CuCl layer. The structure of the (CuCl)/Si superlattice for $n = 3$ and the corresponding Brillouin zone (BZ) with symmetry points are shown in Fig. 2. Remarkably, the superlattice exhibits metallicity at interfaces of the two semiconductors with appreciable band gaps of 3.4 eV (CuCl) and 1.14 eV (Si)[22]. This metallicity is not due to the well-known LDA band gap underestimation, since LDA gives finite band gaps

for both CuCl and Si.

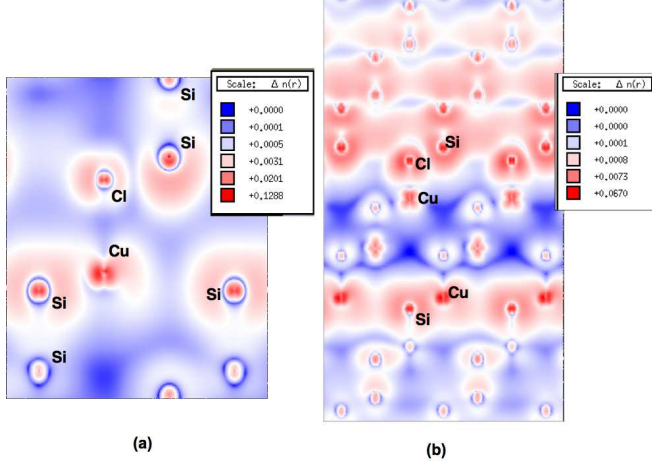


FIG. 3. (Color online) Plot of charge densities at ± 0.25 eV with respect to E_F for $n = 1$ and (b) $n = 3$. Values in iso-plots are in logarithmic scale.

The metallicity at the interface is revealed in the charge density plot as shown in Fig. 3, where an energy slice of ± 0.25 eV with respect to E_F is taken, which correspond to density of $7.3 \times 10^{21} \text{cm}^{-3}$ and $4.3 \times 10^{21} \text{cm}^{-3}$ for $n = 1$ and $n = 3$, respectively. The electronic structure also shows the interfacial metallicity [Fig. 4(a)-(d)], where the n -type (p -type) metallicity is from the Si-Cu (Si-Cl) interface. Furthermore, the Fermi surface (FS) evidences two-dimensionality with little dispersion along the z axis at the zone center for the hole-FS and at the zone corner for the electron-FS. As manifested in the plots of charge densities, bands, and Fermi surfaces, the CuCl/Si superlattices exhibit interfacial metallicity, which can be viewed as the insulator-metal-insulator sandwich structure, as Ginzburg proposed. Hence, the possible superconductivity of this “sandwich” structure is here presented.

Excitonic Superconductivity. In the presence of screening, the screened Coulomb interaction is expressed as[30]

$$V_{\mathbf{G},\mathbf{G}'}^s(\mathbf{q},\omega) = \frac{4\pi e^2}{|\mathbf{q}+\mathbf{G}|^2} \varepsilon_{\mathbf{G},\mathbf{G}'}^{-1}(\mathbf{q},\omega) \quad (2)$$

$$V^s(\mathbf{r},\mathbf{r}';\omega) = \sum_{\mathbf{q}} \sum_{\mathbf{G},\mathbf{G}'} e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}} \frac{4\pi e^2}{|\mathbf{q}+\mathbf{G}|^2} \times \varepsilon_{\mathbf{G},\mathbf{G}'}^{-1}(\mathbf{q},\omega) e^{-i(\mathbf{q}+\mathbf{G}')\cdot\mathbf{r}'} \quad (3)$$

where the inverse dielectric function is, within the random-phase approximation (RPA)

$$\varepsilon_{\mathbf{G},\mathbf{G}'}^{-1}(\mathbf{q},\omega) = \delta_{\mathbf{G},\mathbf{G}'} - \frac{4\pi e^2}{|\mathbf{q}+\mathbf{G}|^2} \chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega), \quad (4)$$

and the generalized susceptibility, $\chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega)$ is given by

$$\chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = \frac{1}{\Omega} \sum_{\mathbf{k}} \sum_{n,m} \frac{f_{\mathbf{k}+\mathbf{q},n} - f_{\mathbf{k},m}}{E_{\mathbf{k}+\mathbf{q},n} - E_{\mathbf{k},m} - \omega + i\eta} \times \langle \mathbf{k} + \mathbf{q}, n | e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}} | \mathbf{k}, m \rangle \langle \mathbf{k}, m | e^{-i(\mathbf{q}+\mathbf{G}')\cdot\mathbf{r}} | \mathbf{k} + \mathbf{q}, n \rangle, \quad (5)$$

where Ω is the volume of the cell, n, m are band indices, $f_{\mathbf{k},n}$ is the Fermi-Dirac function of the n -th band at \mathbf{k} , and \mathbf{G}, \mathbf{G}' are reciprocal lattice vectors; the k summation in Eq. (5) is done over the full BZ.

We emphasize here that it is the inverse dielectric function, $\varepsilon^{-1}(\mathbf{q},\omega)$, rather than the dielectric function, $\varepsilon(\mathbf{q},\omega)$, that determines the screening in Eq. (2). Moreover, it is the inverse dielectric function, $\varepsilon^{-1}(\mathbf{q},\omega)$, that is the true response function for any external perturbation which satisfies the causality relation[31–33], which leads to $0 < \varepsilon^{-1}(\mathbf{q},\omega) < 1$, or $\varepsilon(\mathbf{q},\omega) > 1$ or $\varepsilon(\mathbf{q},\omega) < 0$: The negative sign is allowed for $\varepsilon(\mathbf{q},\omega)$ without violating the stability criterion[31–33].

The pairing interaction is formulated with time-reversal symmetry[34] and considering only singlet pairing, in which a Cooper pair scatters from $(\mathbf{k}, -\mathbf{k})$ to $(\mathbf{k}', -\mathbf{k}')$ in the presence of the screened interaction,

$$V_{\mathbf{k},\mathbf{k}'}^P(\omega) = \langle \mathbf{k}, -\mathbf{k} | V^s(\mathbf{r},\mathbf{r}';\omega) | \mathbf{k}', -\mathbf{k}' \rangle \quad (6)$$

$$= \sum_{\mathbf{G},\mathbf{G}'} \sum_{n,m} \frac{4\pi e^2}{|\mathbf{q}+\mathbf{G}|^2} \varepsilon_{\mathbf{G},\mathbf{G}'}^{-1}(\mathbf{q},\omega) \times M_{n,m}(\mathbf{k},\mathbf{q},\mathbf{G}) \cdot M_{n,m}^*(\mathbf{k},\mathbf{q},\mathbf{G}'),$$

where $M_{n,m}(\mathbf{k},\mathbf{q},\mathbf{G}) = \langle \mathbf{k} + \mathbf{q}, n | e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}} | \mathbf{k}, m \rangle$ is the optical matrix. [For a more detailed derivation, see Supplementary Information.]

It is important to note that having an attractive pairing, $V_{\mathbf{k},\mathbf{k}'}^P < 0$, for particular k points and frequencies is not sufficient to ensure the pairing. Instead, the Kernel function, $K(\omega)$, the average of the pairing interaction over the BZ zone,

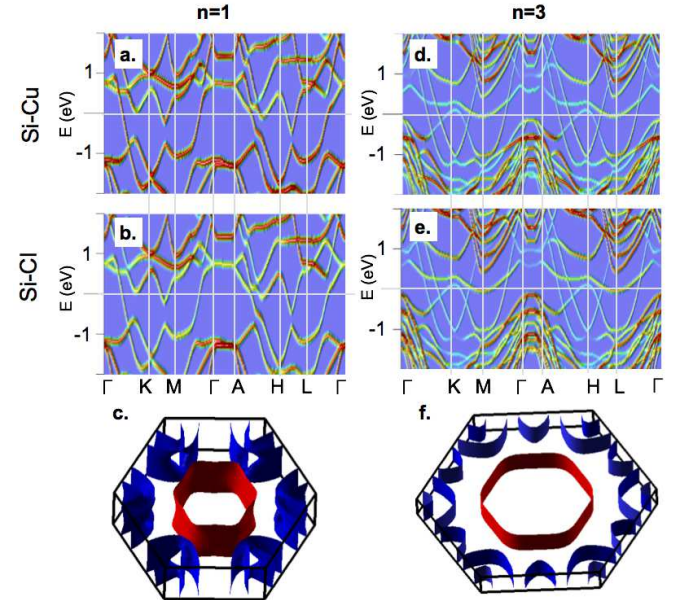


FIG. 4. (Color online) Band structure of the CuCl/Si superlattice for $n = 1$ [(a)-(c)] and $n = 3$ [(d)-(f)]. Contributions from (a),(d) n -type and (b),(e) p -type interface. The Fermi surfaces for (c) $n = 1$ and (f) $n = 3$, where the electron- and hole Fermi surfaces are shown in red and blue, respectively.

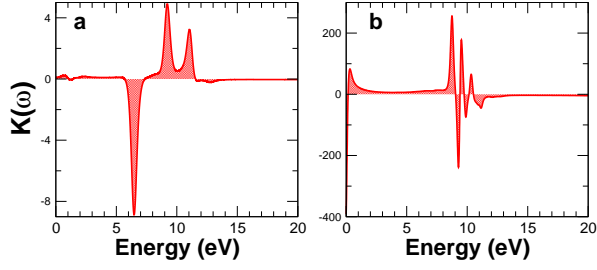


FIG. 5. (Color online) Kernel function, $K(\omega)$, for (a) $n = 1$ and (b) $n = 3$ CuCl/Si superlattice

should be attractive. The concept of the Kernel function was first introduced by Cohen when he proposed superconductivity in doped semiconductors[35] and it is extended in Lithium under high pressure by Akashi and *et al.*[36]. Here we rewrite the Kernel function as

$$K(\omega) = \sum_{\mathbf{q}} K_{\mathbf{q}}(\omega), \quad (7)$$

where $K_{\mathbf{q}}(\omega)$ is \mathbf{q} decomposed Kernel function. [For derivation of the Kernel function, see the Supplementary Information.]

The calculated $K(\omega)$ of the CuCl/Si superlattices are shown in Fig. 5 for $n = 1$ and $n = 3$, respectively. Indeed, regions of $K(\omega) < 0$ are found, a clear indication that pairing can exist due to the electronic interactions alone without involving lattice or spin degrees of freedom. The attractive interaction occurs around 6 eV and 9 eV for $n = 1$ and $n = 3$, respectively. While the attractive region of $n = 1$ is rather wide, that in $n = 3$ is narrow. In both cases, regions of highly repulsive interaction are present nearby the attractive region. However, the presence of the repulsive region of $K(\omega)$ does not necessarily prevent pairing. In phonon mediation, pairing occurs in a very thin range of energy (Θ_D) in the Fermi surface. At least in s -wave pairing symmetry, the presence of attractive region in $K(\omega)$ favors pairing.

T_C as function of λ is plotted in Fig. 6. Θ_E is approximated to 6 and 9 eV from Fig. 5 for $n = 1$ and $n = 3$, respectively. Fig. 6(a) is without Coulomb repulsion ($\mu^* = 0$) using Eq. (1), where phonon pairing with $\Theta_D = 300$ and 600 K are shown for comparison. One notes that T_C by excitonic pairing with $\mu^* = 0$ for $\lambda > 0.5$ become unrealistically high. The case with nonzero μ^* needs some caution. T_C with $\mu^* = 0.1$ is shown in Fig. 6(b), where McMillan's equation is used. As well expected, T_C is reduced with inclusion of μ^* . Even when $\mu^* = 0.2$ [dotted line], enhancement of T_C is evident despite reduction compared to Fig. 6(a).

In this T_C estimate, the validity of Migdal theorem, whether to include higher order vertex correction, is ignored for simplicity. It has been argued that T_C reduces by vertex correction[38]. On the other hand, enhancement of T_C in layered systems has been proposed even with inclusion of vertex correction[39, 40]. As Coulomb repulsion (μ) is renormalized to $1/\mu^* = 1/\mu + \log\left(\frac{\omega_F}{\Theta_D}\right)$ in phonon mediation[41], in

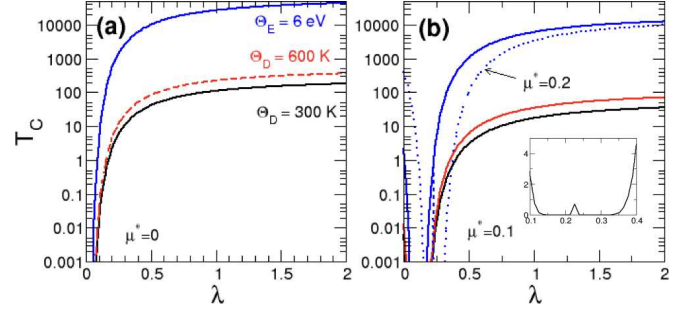


FIG. 6. (Color online) T_C (in logarithmic scale) as function of λ for (a) without Coulomb repulsion using Eq. (1) and (b) with Coulomb repulsion, $\mu^* = 0.1$, using McMillan's equation[37]. T_C with excitonic pairing with $\Theta_E = 6$ eV is shown in blue, phonon pairing with $\Theta_D = 300$ and 600 K are shown in black and red dotted lines, respectively. For simplicity, the asymptotic behavior of T_C for a large value of λ is ignored. The electron-exciton pairing constant, λ , here is defined as done for the electron-phonon coupling constant. The case for $\mu^* = 0.2$ is shown in dotted line in (b). Inset: T_C (in linear scale) for $\mu^* = 0.2$ near λ between 0.1 and 0.4.

the excitonic pairing, Θ_E replaces Θ_D hence μ^* will be larger than phonon mediation. Nevertheless, as shown in Fig. 6, T_C enhancement by exciton mediation is evident[42].

Recall that the diamagnetism of CuCl/Si [111] was reported to be in the 60–150 K range, and calculations within the electron-phonon mediation gave $T \leq 2$ K[22] for this system; thus if CuCl/Si [111] is indeed superconducting, it cannot be solely by phonon mediation. We want to point out here that exciton pairing is not the sole pairing mechanism of purely electronic in character. Kohn and Luttinger[43] suggested that purely electronic pairing due to the sharp edge of the Fermi surface can be realized via a Friedel-like density oscillation, which enables an attractive interaction, whose T_C , however, is too low to be observed experimentally.

Summary In summary, the CuCl/Si superlattice was revisited. It clearly shows metallicity at interfaces, as evidenced in bands, Fermi surfaces and charge density plots. The possibility of excitonic pairing is shown by calculating the Kernel function $K(\omega)$. The attractive effective interaction can exist for frequencies ranging over several eV in favor of pairing with large prefactor in the T_C equation. In spite of renormalization, which would increase μ^* in the exciton pairing, enhancement of T_C over the phonon mediation is evident.

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